

Interactive comment on “Sulfur cycle and sulfate radiative forcing simulated from a coupled global climate-chemistry model” by I.-C. Tsai et al.

Anonymous Referee #1

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Overall Assessment. This paper uses a GCM to simulate the natural and present-day atmospheric sulfur cycle and infer the corresponding direct and indirect radiative forcings and climate responses, including climate feedbacks on sulfate. It adds to the body of global sulfur model studies and provides budgets and radiative forcings that can be compared to the literature. Compared to previous models the direct radiative effect is found to be weak, while the indirect radiative effect is found to be large, but there is no discussion of why. AEROCOM intercomparisons have previously dissected differences in radiative forcings found between models in terms of differences in model processes, and it seems that such an analysis would be useful here. The climate feedback on sulfate is found to be very small and I wonder whether it is significant, considering that a single climate simulation was conducted for each case (no ensemble). The use of fixed SSTs in the climate simulations would be expected to greatly dampen the

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climate response, including in particular the hydrological cycle which affects sulfate. The authors find that the climate response from the direct radiative forcing is too weak to be significant but this could likely be made significant if more model statistics were acquired through ensembles of simulations. The most interesting result of the paper to me was the large sensitivity of the indirect radiative forcing results to the choice of cloud droplet number parameterization. But even that was to be expected. Overall I think that this paper may be publishable after consideration of the comments below, but I don't think that it represents a major contribution.

Detailed comments:

1. Page 4: use of fixed SSTs greatly limits the climate response in the model, including in particular the hydrological cycle, and this should be noted in section 2.1.
2. Page 5, Table 1: the sulfur mechanism presented in Table 1 seems very old. Isn't DMSO now thought to be a major product of DMS oxidation?
3. Table 1: aqueous-phase reactions should not be represented as equilibria.
4. Page 7: I am confused as to whether the N and A simulations differ solely in their sulfur emissions or in their emissions of NO_x, CO, and VOCs as well. At several points the text makes mention of increased ozone in the A simulations due to anthropogenic emissions. However, the negative radiative forcing for A0-N0 (page 14, Table 4) would then not make sense if it's driven by the ozone increase (page 14) since ozone should have a positive radiative forcing.
5. Page 7: doing a single GCM 13-year simulation for each case, with analysis only of the last five years, does not provide very good statistics for climate response. Standard practice is to conduct ensembles of simulations to reduce the effect of noise. This should be at least acknowledged.
6. Page 8: section 3.1 on global distributions of sulfur species is very long and does not contain much that's new.

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7. Page 9: there are actually a number of long-term sulfate data sets in remote air that could be used for model evaluation and these are in fact routinely used by models for that purpose. For example the SEAREX Pacific data come to mind and would be well suited for a model using 1990 emissions.

8. Page 11: I was surprised by the dominant role of ozone as an aqueous-phase SO₂ oxidant. Models generally find H₂O₂ to be the dominant oxidant. At pH = 4.5 (Table 1), oxidation by ozone should be very slow. I am confused by the units given for k_{O3} (and the other aqueous-phase rate constants) in Table 1, as these are units are for rates and not constants. The reaction between SO₃²⁻ and O₃ goes at a rate constant close to the aqueous-phase diffusion limit ($\sim 1.0 \times 10^9 \text{ M}^{-1} \text{ s}^{-1}$), but that's not apparent from Table 1.

9. Page 15, end of 1st paragraph: the argument about robustness seems circular.

10. Page 15: the authors find an anthropogenic sulfate forcing at the low end of the published range for the direct forcing, and at the high end for the indirect forcing. This seems to call for some explanation. The published AEROCOM intercomparisons provide some metrics for attributing model differences in forcings in terms of differences in model parameters.

11. Page 18: a new aspect of this paper is the feedback of climate response on the sulfur cycle, although the use of fixed SSTs limits the response of the hydrological cycle. Page 18 reports very small effects on sulfate: are these effects actually significant (outside interannual variability)?

12. Page 24: climate responses given in the conclusions are greatly limited by the assumption of fixed SSTs and this should be stated.

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